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Adhesion and Cohesion

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Cohesion and adhesion are essential functional properties of certain of the constituents of food mixtures if we wish to convert them to sight-appealing shaped products with acceptable textures. Parker and Taylor (1) define adhesion as the use of one material to bond two other materials together and cohesion as the joining together of the same material. The development of textured foods based on vegetable proteins, multi-component breakfast foods, and some prepared meat specialty items has required at least one ingredient, usually protein, to serve as binder to hold components together. The binding agents may function before or after cooking the ingredient mix; cooking establishes additional adhesive and cohesive interactions among protein, lipid, and carbohydrate components of foods.

In this paper, we will explore the measurement of and the basis for the cohesive and elastic properties of a commonly used component of foods that excels in these characteristics, wheat gluten. Gluten constitutes from 10 to 16% of wheat flour, from which it may be separated by Martin, batter, or Raisio processes (2, 3). The separated wheat gluten is 70 to 80% protein, of which 85% is insoluble in saline solution. We shall also seek to correlate some of the basic concepts developed in studies of gluten to other protein systems, such as those of soybean protein isolates and concentrates.

A good example of the contribution of protein to adhesion and cohesion of a multi-component system is wheat flour dough. Khoo et al. (4) have observed with the scanning electron microscope that dough consists of starch granules held together by a matrix of hydrated gluten protein, which is stretched into coherent films (Figure 1). These films are not artifacts of microscopy since isolated gluten is an excellent film former. Whole gluten purified by solubilization in dilute acids can be dispersed in lactic acid, which acts as a humectant and plasticizer, and cast as a film as shown by Wall and Beckwith (5). Such films would serve as edible coatings. Film-making properties are characteristic of many polymers,

including proteins, and are a good measure of cohesive strength. Gluten proteins have also been tested as industrial adhesives for bonding paper or wood (5). Just as gluten proteins associate with starch, so does it bond to other polar materials, such as cellulose. Many of the concepts developed in industrial adhesives apply to food applications.

Experimental Procedures

Many instruments are available to provide fundamental information on the adhesive and cohesive properties of food products of different forms and at different stages of preparation. Voisey and de Man (6) classify such instruments into two main categories: (a) linear motion instruments which generally measure extensibility and tensile strength or compression and flexing strength, and (b) rotary motion devices which measure resistance to flow or mixing of viscous solutions or plastic masses. The linear motion instruments usually rupture the structures and so are best used to measure a specific stage of product development. In contrast, the rotary instruments often may be used to examine transitory effects on physical properties induced by temperature variation, chemical additives, or stresses caused by mixing.

The most widely used linear motion analyzers are the Instron Universal Testing machines which range from large floor models to table instruments suitable for most food tests (7, 8). Force is applied by drive screws to a crosshead which is moved downward at a specified rate. An upper force measurement cell measures extending forces on materials clamped between it and the moving crosshead (Figure 2). Wall and Beckwith (5) used this system to measure adhesion by wheat gluten in model systems. Data on force and distance traversed by the crossarm are recorded. Frazier et al. (9) used the Instron to compress dough or gluten balls at a given load and measure time required for the balls to relax to a lower load at a constant deformation. Various attachments to the Instron permit measurement of lateral stresses such as flexing. Rasper (10) has modified the Instron to permit dough extension measurements in a temperature-controlled chamber containing fluid with density equal to that of dough. Instruments specifically designed for dough extensibility and tensile strength measurements are the Brabender Extensograph or Simon Extensometer (10). In the extensograph, a rod-shaped mass of dough is clamped horizontally and a hook extends it vertically at a constant rate. Resistance and extension are recorded until the dough cylinder breaks. The area under the curve and the ratio of resistance to extensibility provide useful information as to the elasticity of the dough.

Hydrated food products exhibiting adhesive or cohesive properties are generally highly viscous or plastic and, therefore,

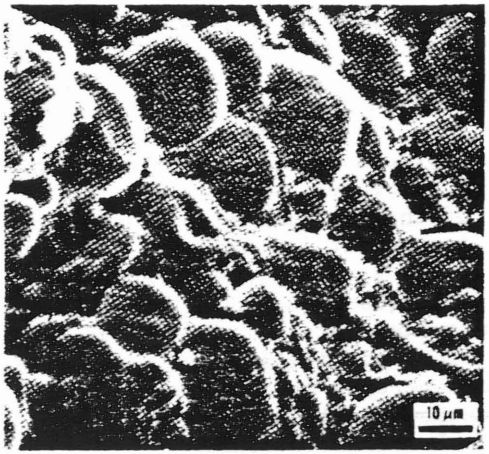


Figure 1. SEM of wheat flour dough after mixing (4)

Figure 2. Tensile strength measurement with the Instron Universal Testing machine

require special rotary instruments for measurement of flow properties. A highly sophisticated instrument designed for elastomer and plastic research but highly useful for research on foods is the Mechanical Spectrometer (Rheometrics Inc., Union, NJ). A considerable variation in shear rates can be applied in a number of different geometries including rotating cone and plate between which a thin layer of sample is inserted. The mechanical assembly and microprocessing unit permit analysis of a broad spectrum of molecular responses yielding data on viscous and elastic properties of the test material. A more generally available laboratory instrument is the Haake Rotovisco viscometer which also features a cone-plate attachment. This modified instrument records torque required to maintain constant rate of rotation and provides data only on the viscous component of flow (8). In the flour milling and baking industries, recording mixers, designated farinographs or mixographs, are widely used for empirical evaluation of flour performance during dough processing. In the Brabender Farinograph, flour and water are mixed by revolving blades. The motor shaft is connected to a dynamometer which measures resistance to the mixing (11, 12). The chart records the dynamic process of dough making and breakdown with time. Water content (absorption) is adjusted to give similar resistances at maximum dough strength. The mixograph is similar in concept to the farinograph but uses pins instead of blades and imposes a greater stress on the dough (11).

While these methods can provide useful information for determining the functional performance of ingredients used in food, the criteria for quality must be established on the final product; thus, dough must be baked into bread and meat analogues cooked and these foods subjected to organoleptic evaluation for texture.

Molecular Interactions

Extensive studies in protein chemistry (13) and synthetic polymers (14) have established compositional factors responsible for molecular associations involved in adhesion and cohesion phenomena of proteins and other polymers. Figure 3 summarizes types of functional groups in proteins participating in associative interactions and agents that disrupt the bonds they form.

Electrostatic charges due to ionized acidic or basic amino acids influence protein solubility. At extremes of pH, many poorly soluble proteins are dissolved and their molecular structures unfolded due to surplus of similar repelling charges. Gluten proteins have few charged groups and so are poorly soluble in neutral solution (15). Dispersions of other proteins must be adjusted to their isoelectric point or have salt added to optimize cohesion and adhesion.

Polar groups contribute greatly to adhesion of proteins to carbohydrates and to their cohesion. In denatured or unfolded proteins, such as animal glues, the peptide amide groups play an important role in adhesion; but in the undenatured collagen, most peptide groups are associated in helical conformations. In undenatured proteins, side chain amide groups from the amino acids glutamine and asparagine and hydroxyl groups of serine and threonine interact through hydrogen bonds. Gluten proteins contain over 33% glutamine and asparagine in their amino acid composition (15).

Investigators of the chemistry of adhesion refer to nonpolar interactions involving long chain aliphatic or aromatic groups in terms of Van der Waal or London forces (1). Protein chemists generally use the term "hydrophobic bonding" to describe these interactions, because in aqueous systems nonpolar residues in proteins tend to retreat and associate in the molecule's interior or with other like groups on adjacent molecules. Membrane proteins especially have exposed hydrophobic groups which contribute to association with lipids and integrity of the membrane structure.

Disulfide bonds in the amino acid cystine are important to the properties of many proteins by maintaining covalent intramolecular bonds and crosslinks between protein chains (16).

The opposing effects of hydrogen bonding and negatively charged amino acid side chains on molecular aggregation of proteins have been demonstrated by experiments with model systems (17). Synthetic polypeptides were prepared containing both polar hydrogen bond-forming glutamine residues and glutamic acid groups. As shown in Figure 4, 2M and 8M urea helped solubilize the polymers in aqueous solutions at low pH by dissociating hydrogen bonds between amide groups. But as the fraction of glutamine residues was increased in the polypeptides, it was necessary to raise the pH to induce more negative electrostatic charges on the glutamic acid residues in order to dissociate the polypeptides held together by hydrogen bonds.

The participation of hydrophobic groups of native wheat gluten proteins in intermolecular associations was demonstrated by Chung and Pomeranz (18) through use of hydrophobic gels as illustrated in Figure 5. These workers introduced a solution of gluten proteins in 0.01M acetic acid into a column of Phenyl-Sepharose-4B. Little protein was eluted by washing the column with dilute acetic acid, but about 40% of the protein was eluted by a solution containing 1% of the detergent sodium sodium dodecyl sulfate. Only a small amount of additional protein was eluted by other solvents including 0.005M glycine-NaOH in 50% propylene glycol. Thus, although hydrophobic bonds are individually weak, their combined strength in proteins, which can provide many such interactions, can be considerable.

The role of noncovalent bonds in determining protein structure and aggregation has been confirmed by x-ray analysis

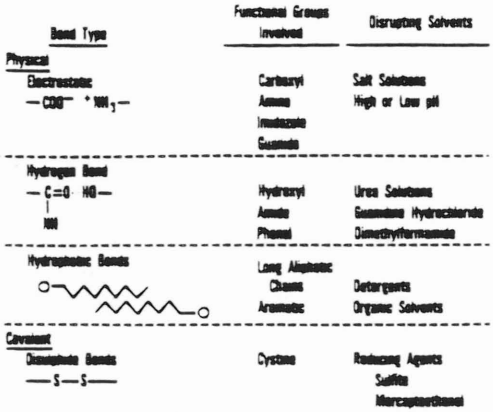
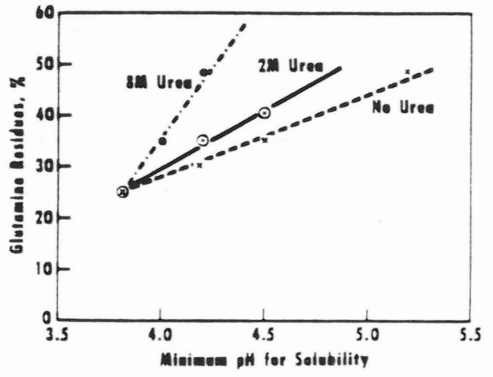


Figure 3. Types of bonds between protein chains



Biochemistry

Figure 4. Influence of urea and pH on the solubilities of synthetic polypeptide copolymers of glutamine and glutamic acid (17)

of protein crystals. At 2Å resolution, the sites of the individual amino acids and the proximities of their functional groups can be deduced, and the nature of the bonding forces involved in maintaining the folding and association of polypeptide chains can be established. Studies on the structure of the molecules of the enzyme trypsin, the soybean trypsin inhibitor, and the complex between the two proteins indicate that multiple types of noncovalent linkages involving hydrogen bonds, hydrophobic bonds, and electrostatic attractions participate in the molecular association (19).

Molecular Size and Shape

Not only do the kinds and amounts of functional groups on proteins govern the extent of protein interactions, but their location on the molecule and their accessability to groups in other molecules are important also. These factors depend on the size and shape of the protein molecules. Polymer and adhesive chemists concur that highly cohesive films and other structures are attained by high-molecular-weight molecules that allow extensive molecular interactions and by those with numerous intermolecular covalent crosslinks (1, 14). In contrast, adhesion depends more on accessibility of functional groups of the adhesive to the adhering materials.

As shown in Figure 6, proteins of the gluten complex can be separated by solubility differences into saline-soluble albumins and globulins, 70% ethanol-soluble gliadins, acetic acid-soluble glutenin, and an insoluble protein residue. Albumins and globulins are highly folded compact molecules. Much of their backbone chain is involved in helical hydrogen-bonded associations. Folding is maintained by both internal hydrogen and hydrophobic bonds as well as by disulfide bonds. The gliadins, with few charged groups, associate to yield a syrupy mass on hydration. But the large asymmetric glutenin molecules form a tough, rubbery, cohesive mass when hydrated. The large size of soluble glutenin molecules is due to limited disulfide bonds between polypeptide chains. The insolubility of residue protein is attributable to extensive intermolecular disulfide crosslinks.

Evidence for variation in molecular size of gluten proteins was obtained when protein extracts of flour were chromatographed on agarose gel filtration columns (Sephadex C1-4B) in tris buffer containing sodium dodecyl sulfate as shown in Figure 7 (20). Most albumins (Alb) and globulins (Glob) as well as gliadins elute late, indicating they have molecular weights below 40,000. In contrast, glutenin shows a broad spectrum of species with some components having molecular weight over 1 million. Use of the hydrophobic bond-breaking solvent sodium dodecyl sulfate disrupted aggregation of the proteins. The

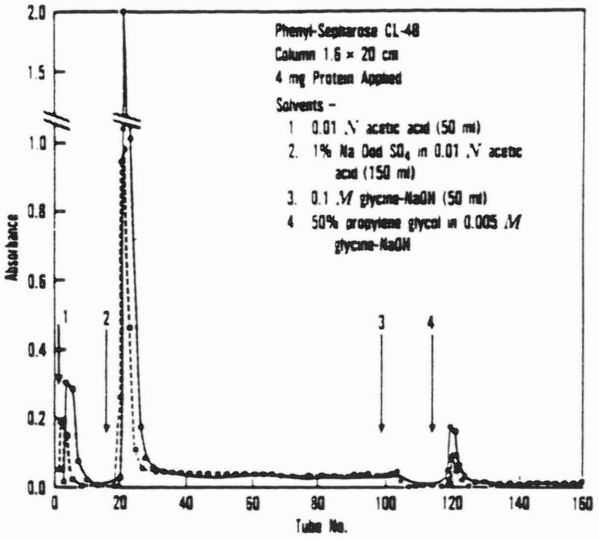


Figure 5. Elution of acid-soluble wheat proteins from hydrophobic gel Phenyl-Sepharose-4B by different solvents. NaDodSO$_4$ = SDS (18).

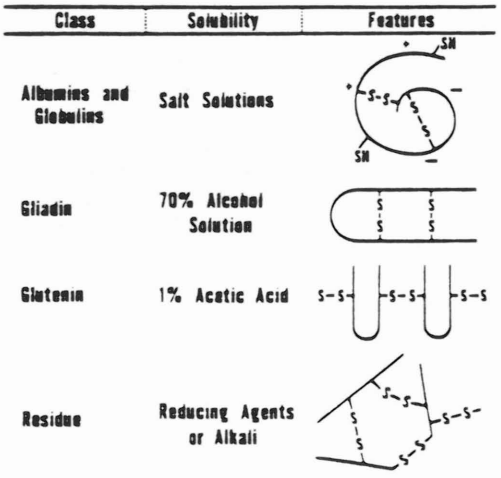


Figure 6. Types of proteins in wheat flour as separated by solubility

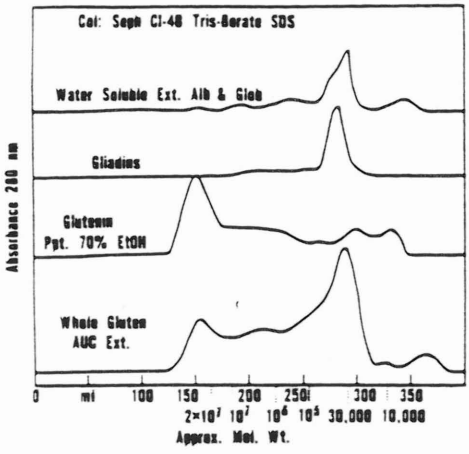


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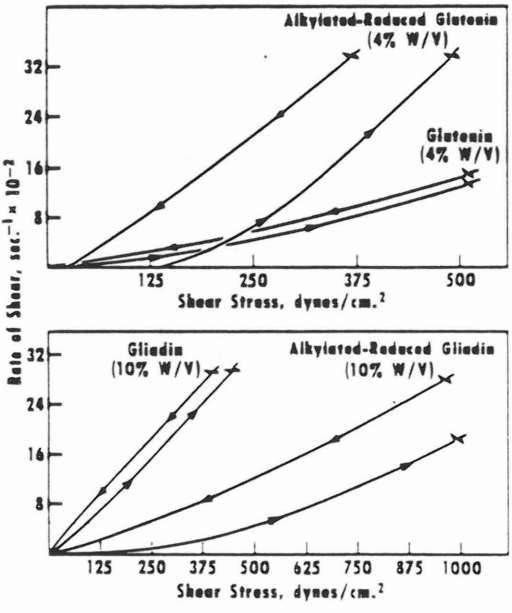
Figure 7. Fractionation of wheat proteins on sepharose-CL4B columns by gel filtration chromatography in 0.125 tris-borate buffer, pH 8.9 and 0.1% SDS (20). AUC ext. refers to protein extracted from wheat flour with a solution containing 0.1M acetic acid, 3M urea, and 0.01M cetyltrimethyl-ammonium-bromide.



Cereal Science Today

Figure 8. Shear–stress curves for native and alkylated–reduced gliadin and glutenin solutions at low shear stresses (5)

low-molecular-weight glutenin protein fraction was not separated in other solvents and appears to consist of membrane proteins that tend to aggregate.

The effect of molecular size and shape on protein cohesive strength was indicated by measurements of tensile strength and elongation of films cast from laboratory preparations of wheat gluten, gliadin, and glutenin (5). The measurements were made with a Scott Tester Model IP-2 at 20°C and 42% relative humidity. Values for force applied to the film (tensile strength) and elongation at time of film rupture are listed in Table I. Glutenin, which consists of the larger, more asymmetric molecules, forms films with greater tensile strength than gliadin films. Gliadin films stretch further than those from glutenin due to their having weaker molecular associations. Gluten, which is a mixture of gliadin and glutenin, has intermediate film properties.

Marked changes in the viscosities of glutenin and gliadin solutions, as measured by cone-plate viscometer, occur after cleavage of their disulfide bonds by a reducing agent (Figure 8). Native glutenin has high viscosity, which indicates not only high molecular weight but also a highly asymmetric structure (5). Furthermore the hysteresis or deviation of the viscosity curves for increasing and decreasing shear stress versus rate of shear provide evidence for non-Newtonian behavior of these molecules due to molecular interactions. Glutenin viscosity

TABLE I

Tensile Strength and Percent Elongation of Wheat Protein Films

Film	Tensile strength $lb/in^2 \times 10^{-3}$	Elongation %
Glutenin	3.38	63
Gliadin	1.42	72
Whole gluten (laboratory preparation)	1.75	75

From Wall and Beckwith (5).

declines markedly when its intermolecular disulfide bonds are cleaved to liberate its constituent polypeptide chains. The viscosity of native gliadin in solution is much less than that of glutenin. Reduction of the disulfides of gliadin results in a significant increase in its viscosity due to unfolding of the polypeptide molecule. Reduction of glutenin destroys its cohesive nature when hydrated, but the reduced proteins are very sticky and quite adhesive.

Dough Rheology

Because of the gluten proteins, hydrated flour can be worked into an elastic-cohesive mass by mixing. The development of optimal dough properties with time during the mixing process can be followed on a mixograph. The mixograph recordings in Figure 9 show that initially the unoriented dough molecules offer little resistance. As mixing proceeds, the asymmetric glutenin molecules are oriented and associate to increase dough strength. Disulfide-sulfhydryl interchanges to facilitate rearrangement of the molecule and actual cleavage of the disulfide links may also take place during mixing (23). Finally, resistance to mixing declines as polymer disruption continues. The three curves show different mixing responses from flours of wheats of different breadmaking quality as measured by Finney et al. (21) and Finney and Shogren (22). The middle curve shows resistance changes with time for dough from a flour with suitable properties for breadmaking. It exhibits a moderate dough development time and stability time. The upper curve is that for dough from a wheat whose gluten is overly strong, since it requires longer mixing to achieve maximum dough strength, whereas the lower curve is of a weak flour dough with short mixing time requirement but rapid breakdown of dough strength.

The relationship between different flours varying in dough strength and their composition of different protein fractions was investigated by Orth and Bushuk (24) and by Huebner and Wall (25). The former workers found a correlation between the mixing time requirement of doughs and their tolerance to mixing to their content of residue protein. As shown in Figure 10, the latter workers analyzed for protein content a series of flours derived from different Hard Red Winter wheat varieties that vary in mixing time requirement (mixing strength). The stronger flours contained not only more residue protein but also more of the higher molecular weight glutenin fraction (Glutenin I).

Changes occurring in disulfide bonds of wheat gluten during dough mixing are supported by two observations. Mecham and Knapp (26) found that mixing in the absence of air increases the content of sulfhydryl groups in the dough. Also, there is an increase in extractable protein during mixing. This increase in extractable protein is primarily high-molecular-weight

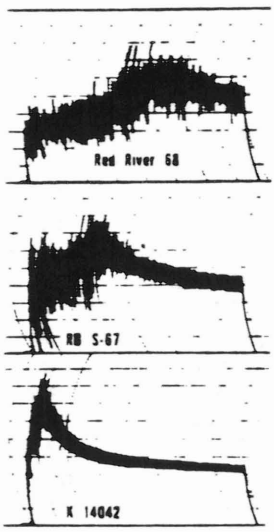
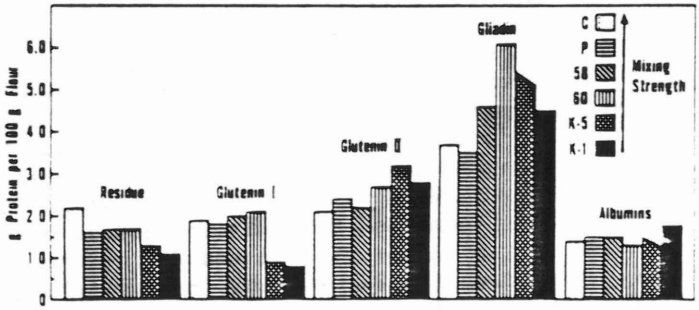


Figure 9. Mixograms of doughs from flours prepared from three varieties of wheats varying in dough performance (21, 22)



Cereal Chemistry

Figure 10. Yields of protein fractions isolated from hard red winter wheat flours differing in mixing strengths (25)

protein according to gel filtration studies conducted by Tsen (27), whose data is given in Figure 11. Apparently, mixing breaks down disulfide bonds in the highly crosslinked residue protein, thereby decreasing resistance to mixing. Addition of reducing agents such as cysteine which cleave disulfide bonds weakens the dough (21), whereas addition of oxidizing agents such as bromate tends to strengthen it by eliminating sulfhydryl groups (26).

Figure 12 summarizes the contribution of various gluten proteins to dough properties. The large asymmetric glutenin molecules have considerable surfaces with numerous exposed functional groups to permit strong association by noncovalent forces. Fragments of highly crosslinked residue proteins contribute lateral cohesion and resistance to laminar flow. During mixing the residue proteins are probably degraded to yield linear, more soluble molecules. The smaller gliadin molecules are less tightly bound and facilitate fluidity and expansion of the dough. These ideas are consistent with experimental findings by Hoseney et al. (28), who separated the gluten proteins from flours of different baking quality and substituted gliadin or glutenin protein from good baking wheat flours for the same protein fraction in poor flours in reconstituted doughs. The gliadin fraction from good wheat flours appeared to improve loaf volume, while the glutenin fraction affected the mixing requirement and tolerance. All components of dough must be present in proper amounts for good breadmaking properties. If the protein is too cohesive and tough, the dough will not rise properly because expansion of trapped yeast-generated CO_2 bubbles will be minimal; but, if the protein matrix is weak, the gas pockets will break and the dough will collapse.

Uses of Wheat Gluten

The unique properties of wheat gluten proteins have resulted in considerable use of isolated gluten preparations. Of the 20 million kg used in the United States, 69% is used in bakery products, 12% in breakfast foods, 9% in pet foods, and 4% in meat analogs (2). In baked goods, gluten may be used to supplement weak flours to provide additional mixing strength and tolerance. It may be used in specialty products, such as high-fiber breads, where the added gluten provides better loaf volume. A major use is in production of hamburger buns, where the supplemented gluten increases the structural strength of the hinge. When hydrated gluten is heated above 85°C, the protein is denatured but retains its shape and its resiliency (29). In bread and rolls, the gluten helps retain moisture in the crumb and contributes to crumb strength.

Gluten is used in many other foods where its adhesive and cohesive properties provide beneficial value (29). In breakfast

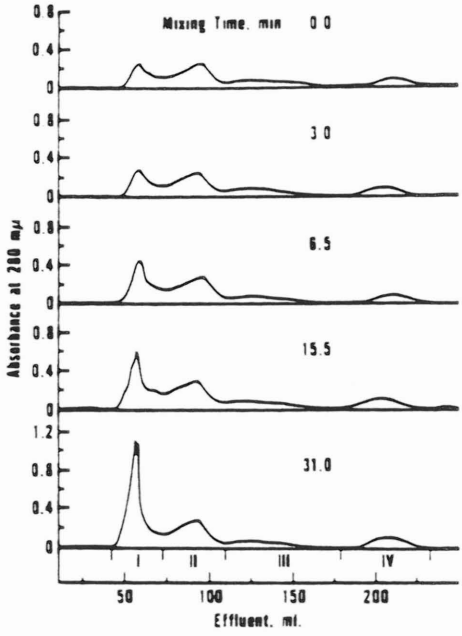


Figure 11. Gel filtration of acetic acid extracts of flour and of dough mixed different times (27)

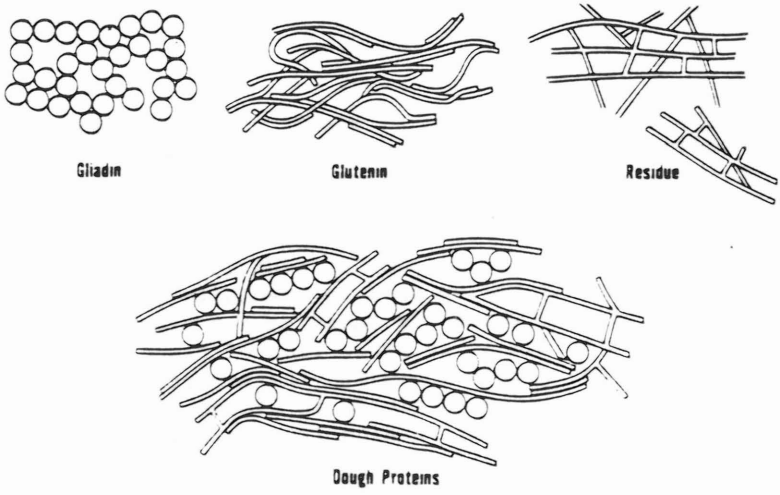


Figure 12. Effect of wheat protein structures on molecular associations and viscoelastic properties

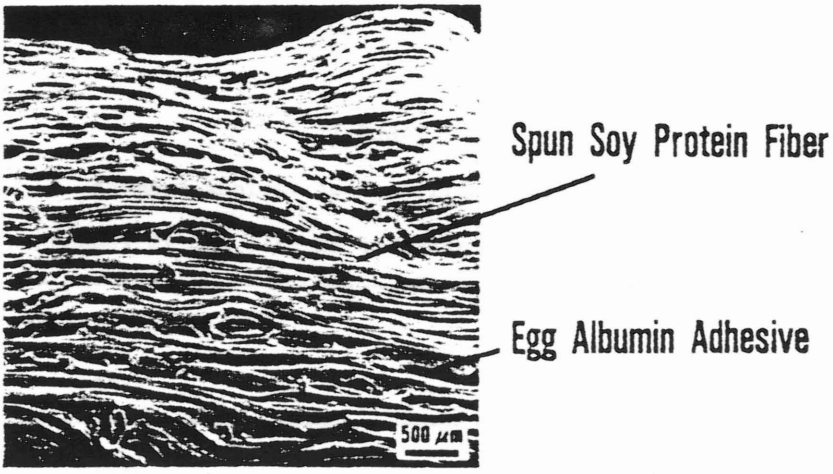


Figure 13. SEM of Bacos spun soybean protein fiber simulated meat product. Egg albumin used as adhesive for fibers (33).

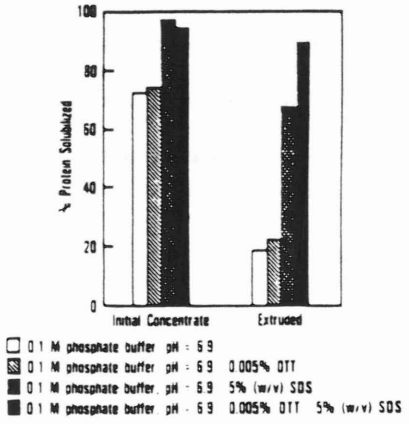


Figure 14. Protein solubility of initial and extruded field bean protein concentrates in different extraction solutions (26)

TABLE II

Meat Binding Abilities of Various Nonmeat Proteins in the Presence of 8% Salt and 2% Phosphate

Protein	Binding ability, grams
Wheat gluten	175.4
Egg white	120.3
Control	107.0
Calcium reduced dried skim milk	74.5
Bovine blood plasma	71.9
Isolated soy protein	66.7
Sodium caseinate	0

Cooking and texturizing vegetable proteins appears desirable if their properties for use in frankfurters or other protein-supplemented meat products are to be optimized. Uncooked soybean or cottonseed proteins do not maintain the texture of prepared frankfurters when added at 10% or 30% levels to the formulations according to Terrell et al. (35). Stress deformation of frankfurters was measured on the Universal Instron Testing machine with a L.E.E.-Kramer press. All-meat frankfurters showed higher values of stress-deformation prior to rupture than products with added soy flour, soy concentrate, liquid-cyclone-processed cottonseed flour, or isolated cottonseed protein. The textured soy flour and textured cottonseed protein performed better than the uncooked proteins; at the 30% level, the addition of textured soy proteins resulted in frankfurters having a higher deformation strength than the all-meat one.

Conclusions

Adhesion and cohesion are properties of many polymeric substances including proteins. The effectiveness of the proteins in bonding or shaping food ingredients is dependent on their composition and structure. Hydrophobic and hydrogen bonding functional groups on amino acids associate with like groups within the protein to influence conformation or between molecules to result in aggregation. Disulfide bonds between proteins result in larger molecules or insoluble complexes. High molecular weight and random coil structure of protein result in more associations and thereby enhance adhesive and cohesive properties. Although these characteristics are inherent in native gluten proteins, functional properties of other proteins may be improved by chemical or thermal processing. There is no universally good adhesive for food constituents. Proteins that are highly cohesive may not blend well with certain other ingredients. It is necessary to examine the available proteins for optimum properties and to select the most satisfactory ingredient combinations. A number of instruments are available for measurements of textural properties of food ingredients or products, but the final criteria for acceptable performance must be taste-panel evaluations.

The mention of firm names or trade products does not imply that they are endorsed by the U.S. Department of Agriculture over other firms or similar products not mentioned.

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